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A simple model of mixing and chemical reaction in a turbulent shear layer

By J. E. BROADWELL AND R. E. BREIDENTHAL†
Graduate Aeronautical Laboratories, California Institute of Technology, Pasadena, CA 91125

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Arguments are presented to show that the concept of gradient diffusion is inapplicable to mixing in turbulent shear layers. A new model is proposed for treating molecular mixing and chemical reaction in such flows at high Reynolds number. It is based upon the experimental observations that revealed the presence of coherent structures and that showed that fluid elements from the two streams are distributed unmixed throughout the layer by large-scale inviscid motions. The model incorporates features of the strained flame model and makes use of the Kolmogorov cascade in scales. Several model predictions differ markedly from those of diffusion models and suggest experiments for testing the two approaches.

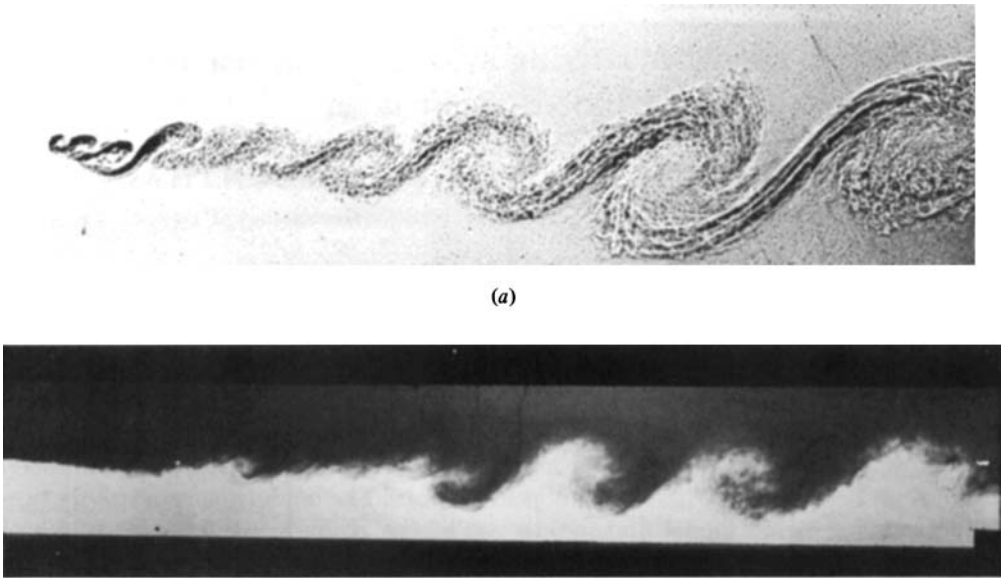
1. Introduction

Recent experimental studies of mixing and chemical reactions in turbulent shear layers and jets provide almost-conclusive evidence that the concept of gradient diffusion is inapplicable to these flows. A part of the evidence is discussed in detail in this section.

As a preface, it should be noted that Corrsin (1974) pointed out several years ago that a necessary condition for a self-consistent gradient-diffusion model is that turbulent motions have a scale that is small compared with that over which the concentration of the diffusing quantity changes significantly and that '...nearly all traditional turbulent transport problems violate this requirement...'. Williams & Libby (1980) more recently specifically urge caution in the use of the concept in combustion analysis. It is irrelevant to this point that the scales that are too large to permit a gradient-diffusion model now appear to be associated with coherent structures. The existence and form of these structures, however, suggest an alternative description of the mixing and reaction processes in such flows. This paper presents a model incorporating these ideas in their simplest form for the two-dimensional turbulent shear layer between two streams of equal, constant density.

We first review the evidence that the path by which the fluids of the two streams reach molecularly mixed states does not resemble the path implied by a diffusive process. As a beginning, consider the shadowgraph in figure 1 (*a*) from the investigation by Konrad (1976) of the low-Mach-number two-dimensional shear layer between gases of various densities. In the side view shown, the gases are of equal density, the velocity ratio is 0·38, and the Reynolds number is $1{\cdot}7 \times 10^4$ cm^{-1}. This shadowgraph and many others from the study, as well as those of Brown & Roshko (1974), show that gases from both sides of the layer are transported across the layer, unmixed, by large-scale motions. Similar motions in water can be seen in the photographs of Breidenthal (1978, 1982).

† Present address: Dept of Aeronautics, University of Washington, Seattle, WA 98195.



(*a*)

(*b*)

FIGURE 1. Turbulent shear layer. (*a*) Gas-layer shadowgraph: upper stream, nitrogen, $U_1 = 1000$ cm/s; lower stream, helium–argon, $U_2 = 380$ cm/s; pressure = 4 atm; channel width = 5·01 cm (from Konrad 1976). (*b*) Water-layer photograph: upper stream, alkaline with phenolphthalein, $U_1 = 70$ cm/s; lower stream, acidic, $U_2 = 40$ cm/s; channel width = 7 cm; equivalence ratio = 1/50.

Before presenting Konrad's concentration measurements, which support this observation, we discuss next the photograph in figure 1 (*b*). Here the shear layer lies between the upper alkaline water stream carrying a visible reactant, phenolphthalein, and the lower stream carrying a colourless sulphuric acid. The Reynolds number is approximately 5×10^3 cm^{-1}, and the equivalence ratio is approximately 1/50, with an excess of the invisible acid. Equivalence ratio is defined here as the volume ratio of acid to alkali fluids required to form the colourless product. That is, the reactant concentrations are such that when only $\frac{1}{50}$ parts of fluid from the lower stream are united with one part of the upper stream, the colour disappears in a fast chemical reaction. The photograph shows, therefore, that tongues of upper stream fluid reach the opposite side of the layer before any significant molecular mixing occurs. At this equivalence ratio, traditional turbulent-diffusion analysis would predict that the reaction takes place along the upper edge of the mixing layer, and that therefore the entire layer would be colourless. The inconsistency with the photograph is evident. (This experiment was done in the apparatus described in Breidenthal 1978.)

Time- and space-resolved concentration measurements have been made by Brown & Roshko (1974), Rebollo (1973), and by Konrad, using the probe described in Brown & Rebollo (1972). Some of Konrad's results, again for the equal-density case (nitrogen on one side and a mixture of argon and helium on the other), are given in figure 2. Figure 2 (*a*) shows the large-scale fluctuations and the presence of unmixed gases that are recorded as the probe is moved slowly across the layer. Even more instructive are the data presented in a different form in figure 2 (*b*). The curves marked $(C(N_2))_I$ and $(C(N_2))_T$ indicate respectively the time-averaged concentration of pure (within 3%) nitrogen and of nitrogen mixed to a greater degree. The remarkable feature of this latter curve, as noted by Konrad, is its near-flatness, i.e. the virtual independence

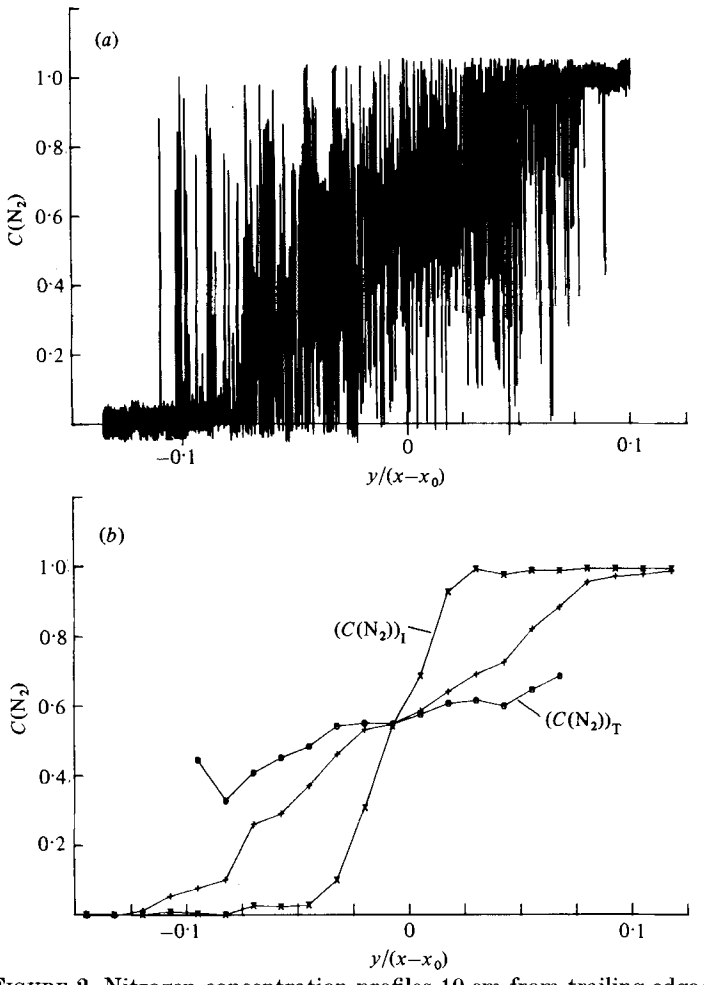


FIGURE 2. Nitrogen-concentration profiles 10 cm from trailing edge; same conditions as for figure 1 (*a*) (from Konrad 1976).

of the mixed-fluid concentration from the lateral coordinate. Fiedler's (1974) measurements of temperature in a mixing layer, one side of which was slightly heated, exhibit similar characteristics. Indeed, inside the vortices the small gradients that do exist are larger in the axial direction!

Scepticism has been expressed (e.g. by Chandrasada *et al.* 1978) that coherent structures, and hence the associated mixing characteristics, will persist to high Reynolds numbers. This issue has been addressed by Dimotakis & Brown (1976), who studied shear layers, generated in a water channel, at Reynolds number of the order of 10^6. The reader is referred to this paper, where several colour photographs of the flow are reproduced. We quote from the paper: 'Dye that labels fluid from one side can be seen to cross all the way to the other side before any small-scale mixing occurs.' Further reference to this investigation is made after the proposed new model has been introduced.

It should be emphasized at this point that in the studies referred to in the preceding paragraphs the time-average profiles of concentration, velocity, and so forth, are in agreement with those that have been known for many years. The structures are revealed only by measurements with sufficient temporal and spatial resolution.

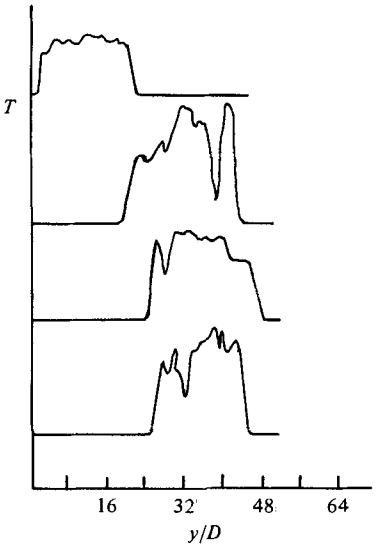


FIGURE 3. Instantaneous temperature profiles across two-dimensional jet at $x/D = 4{\cdot}5$; mean velocity on axis = 0·305 m/s (from Uberoi & Singh 1975).

While not directly relevant to the present study of shear layers, the hot-wire measurements of Uberoi & Singh (1975) in a slightly heated two-dimensional turbulent jet suggest that similar, non-diffusive, processes control the mixing in that flow as well. They projected hot wires across the jet at several axial locations, and were able, in this manner, to obtain virtually instantaneous temperature distributions, examples of which are given in figure 3. Within the jet there is, again, little average lateral temperature variation. These authors interpret their results to mean that the lateral position of the jet is fluctuating, and point out that a superposition of the instantaneous profiles yields the well-known bell-shaped curves.

It could perhaps be argued that, since analyses based on turbulent-diffusion notions have been used successfully to describe the velocity distributions in jets, shear layers, and wakes, the approach may work for molecular mixing. (Many theories are useful, of course, in situations where they cannot be rigorously justified.) That this is not the case is shown by the careful study of Walker (1979). He compared predictions of several commonly used turbulent mixing models with measurements in turbulent jets and mixing layers. Most attention was given to the $k\epsilon 2$ model, developed by Spalding and coworkers (see Dash, Pergament & Thorpe 1979) and the $k\omega'$ model of Saffman and coworkers, described in detail by Walker (1979).

He found that for shear layers of uniform density the velocity-profile predictions of both models were in excellent accord with the experiments of Brown & Roshko (1974). When chemical reactions were considered, however, the outcome was different. Wallace & Brown (1979) measured the temperature distributions in nitrogen shear layers carrying ozone and nitric oxide in low concentrations. The reaction $O_3 + NO \rightarrow NO_2 + O_2$ is spontaneous and calculated to be diffusion-limited. Figure 4 compares the experimental values with those of the $k\omega'$ model, which Walker (1979) extended to treat chemical reactions. Both peak and integral values are seen to differ by a factor of approximately two. Walker presents comparisons with predictions of several other models and finds similar large discrepancies. He concludes that the vortex structure is a crucial missing element in the models.

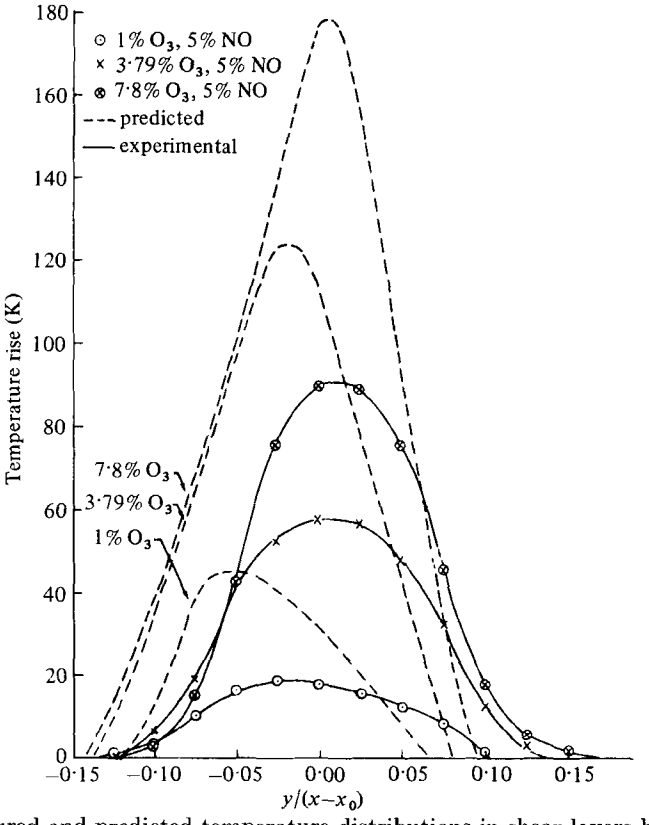


FIGURE 4. Measured and predicted temperature distributions in shear layers between streams carrying nitric oxide and ozone (from Walker 1979).

2. The model

To describe the model that the data discussed above suggest to us, it is convenient to work in a coordinate system moving in the axial direction with a velocity approximately the mean of the two streams. Coles (1981) conjectures, from results such as those in figure 1 (*a*) and from the numerical calculations of Patnaik, Sherman & Corcos (1976) and Corcos & Sherman (1976), that the mean particle paths in this system are as depicted in figure 5. In this figure the upper stream carries a reactant A and the lower, a reactant B. A product C is considered to be formed by an irreversible reaction that is fast compared with the mixing rates. The density is taken to be constant. The interfacial zone into which the reactants diffuse by molecular motion is shown as a cross-hatched layer. The dotted spaces denote regions where the interfacial zonal volume has grown to such large values that the edges have touched.

We are considering flows in which the laminar-diffusion-zone thickness is small compared with the turbulent-layer thickness. This will be the case when two conditions are met: (i) high Reynolds number, $Re \equiv \Delta UL/\nu$, where ΔU is the velocity difference across the layer, L the thickness and ν the kinematic viscosity; (ii) large values of $\Delta UL/D = Re\,Sc$, where D is the molecular diffusion coefficient and $Sc = \nu/D$ is the Schmidt number.

The sketch ignores vortex pairing or amalgamation, which is known to occur in

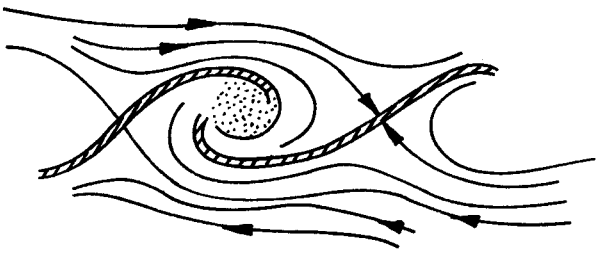


FIGURE 5. Sketch of mean streamlines and molecular mixing zones in shear layer (streamlines from Coles 1981).

this flow. While such events have important dynamical consequences, it is not yet clear what role they play in the subsequent molecular mixing. For this reason, and because, as will be seen, the model is not based on details of the flow field, there is no explicit treatment of pairing.

It is clear from shadowgraphs such as that in figure 1 (*a*) that a given section of interface grows as it is distorted by the secondary motions in the braids between the vortices, and more and more rapidly as it is drawn into the vortices themselves where successively finer scales develop. It is sufficient for the development of the model to argue from the photographic evidence, as well as on intuitive grounds, that the interfacial area S per unit volume, being inversely proportional to the scale λ of the distortions, grows slowly at first and then extremely rapidly as λ approaches its minimum value λ_0, the Kolmogorov scale. (Here we are following in time the behaviour of given elements of reactant and the associated interface.) If, however, we assume that the Kolmogorov description of the cascade in scales applies in the shear layer, at least at the small scales, we can make a more quantitative statement about the growth rate of $S(\sim 1/\lambda)$.

The central idea (see e.g. Landau & Lifshitz 1959) is that for scales down to λ_0 the motion is inviscid and the kinetic energy flux e is constant, i.e. that

$$e \sim \frac{v_\lambda^3}{\lambda} \sim \frac{\Delta U^3}{L} \quad (\lambda_0 < \lambda < L). \tag{1}$$

This statement alone is not sufficient to determine the dependence of λ on the time t, and further progress must rely solely on dimensional arguments. To quote Landau & Lifshitz 'v_λ may also be regarded as the velocity of turbulent eddies of size λ'. Thus, if we assume that in one 'turnover time' the eddy gives up a large fraction of its energy, the time spent at λ_1 is $\Delta t_1 \sim \lambda_1/v_1 \sim \lambda_1/e^{\frac{1}{3}}\lambda_1^{\frac{1}{3}} \sim \lambda_1^{\frac{2}{3}}/e^{\frac{1}{3}}$. Now a further decision is required concerning the new scale generated. V. Kulkarny (private communication) has pointed out that the requirement of self-similarity suggests

$$dt = \kappa \frac{\lambda^{\frac{2}{3}} d\lambda}{e^{\frac{1}{3}}\lambda}, \tag{2}$$

where κ is a constant. (Here we have, for convenience, assumed a continuous change in scale. The actual situation may be better described by jumps between discrete scales, but, as has already been pointed out, we need only qualitative results at this stage.) Integration of (2) and use of (1) give the time to reach a given scale λ, starting at scale L,

$$t(\lambda) = \kappa_1 \frac{L}{\Delta U}\left[1 - \left(\frac{\lambda}{L}\right)^{\frac{2}{3}}\right], \tag{3}$$

and, since $\lambda_0 = \kappa_2 L/Re^{\frac{3}{4}}$,

$$t_\kappa = \kappa_1 \frac{L}{\Delta U}\left[1-\frac{\kappa_2}{Re^{\frac{1}{2}}}\right] \approx \kappa_1 \frac{L}{\Delta U} \quad \text{for} \quad Re \gg 1, \tag{4}$$

where κ_1 and κ_2 are constants and t_κ is the time to reach the Kolmogorov scale. Therefore, at large Reynolds number the time to reach the smallest scale is proportional to $L/\Delta U$, essentially independent of Re. With $S \sim 1/\lambda$ we have for the growth of the surface area per unit volume

$$S \sim \frac{1}{L}\left(1-\frac{t}{T}\right)^{-\frac{3}{2}}, \quad S < \frac{1}{\lambda_0}. \tag{5}$$

The formulation thus predicts a rapid increase in interfacial surface area as the Kolmogorov scale is approached and that at high Reynolds number the time to reach this scale is virtually independent of Reynolds number.

Next we wish to show that, when the scale λ_0 is reached and the interfacial zones begin to overlap, the entire volume in which this occurs is brought quickly to a homogeneous composition by molecular diffusion. (We are for the moment not considering the chemical reaction.) For this purpose we note that the time to diffuse across the Kolmogorov scale is

$$t_\lambda \sim \frac{\lambda_0^2}{D} \sim \frac{L}{\Delta U}\frac{Sc}{Re^{\frac{1}{2}}} \sim t_\kappa \frac{Sc}{Re^{\frac{1}{2}}}, \tag{6}$$

where again D is the diffusion coefficient and Sc the Schmidt number. When Sc is greater than unity, as it is in water, the diffusion-zone thickness (the flame-sheet thickness when reactions occur) is less than the Kolmogorov scale. In this case the (slow) motions that continue at scales smaller than λ_0 aid in the molecular mixing by continued stirring of the diffusion zones. The estimate in (6) thus represents an upper limit for the time required for the region to reach homogeneity. In any event, the important conclusion is that at high Reynolds number the regions of scale λ_0 are made uniform by diffusion in a time that is negligible compared with the time t_κ for the fluid elements to reach this scale.

Specific evidence bearing on the rapidity of mixing at the small scales was obtained in another phase of the Dimotakis & Brown (1976) investigation quoted earlier. Again we paraphrase their remarks and refer the reader to their paper, where the pertinent colour photographs are reproduced. 'To investigate this final state of the mixing process more directly and to obtain a quantitative feeling for the rapidity of the subsequent molecular mixing, a different scheme was employed.' Diluted nitric acid containing bromothymol blue, a pH indicator, was introduced into one stream, and colourless ammonium hydroxide solution in the other. The bromothymol blue changes from deep orange in acidic environment to deep blue in the alkali, the change occurring close to a pH of 7. Concerning the photograph, they state 'The acid in the picture (orange) has already entered the shear layer. . . and can be seen to participate in the large structure's motion. It does not react (become blue), however, until later. It can be seen that, at these Reynolds numbers, the reaction, once it begins, occurs almost instantaneously. . . . Thus it would appear that the processes of entrainment and fine-scale mixing, at high Reynolds number, are almost distinct and separate stages.' The flame-sheet-area dependence on time in the Kolmogorov cascade, and the subsequent rapid diffusion would seem to be in accord with these observations.

The experimental evidence cited in §1 and in the preceding paragraphs, the rapid

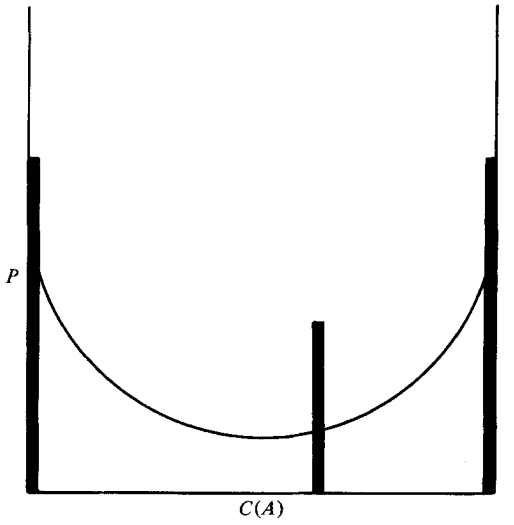


FIGURE 6. Sketch of model probability density function.

increase in S as $\lambda \to \lambda_0$, and the existence of the two widely separated scales t_κ and t_λ suggest the following idealization for the fluid states in the turbulent shear layer. The fluid in the layer is in three states: (i) unmixed, i.e. pure fluids from the two streams, (ii) a homogeneous mixture at the entrainment ratio, and (iii) in strained laminar diffusion layers between the free-stream fluids.

The model is, of course, an oversimplification. For instance, figures 1 and 5 suggest that parts of the interdiffusion zones are bounded on one side by the homogeneous region instead of the free streams containing only A and B. This different boundary condition not only introduces different compositions into the diffusion zone but also implies that all of the scale changes may not follow the cascade described above. Some fluid may be directly entrained into the smaller scales. Nevertheless it seems inappropriate to introduce refinements to account for such effects before more evidence is available to support the basic notions being put forward.

To describe the probability-density distribution that the model in its present form implies, we need a description of the laminar-diffusion-zone structure. If we assume that the strain rate adjusts sufficiently rapidly to the changing scales, i.e. that the structure is quasi-steady and that the zones may be considered plane, then the analysis of Marble (described e.g. in Marble & Broadwell 1977) is applicable. The distributions of A and B (still considered to be inert) within the zones are

$$\frac{\kappa_A}{\kappa_A(\infty)} = \frac{1}{2}\left\{\mathrm{erf}\left[\frac{y}{(D/2\epsilon)^{\frac{1}{2}}}\right]+1\right\}, \tag{7}$$

$$\frac{\kappa_B}{\kappa_B(\infty)} = \frac{1}{2}\left\{\mathrm{erf}\left[\frac{-y}{(D/2\epsilon)^{\frac{1}{2}}}\right]+1\right\}, \tag{8}$$

in which y is the coordinate normal to the zone, ϵ is the local strain rate, and (∞) denotes conditions far from the layer. These error functions yield a U-shaped p.d.f. The regions of pure A and pure B are delta functions at the extemities, and the homogeneously mixed region a delta function at the entrainment ratio E. (Konrad shows that E is about 1·3 at the velocity ratio 0·38 that he studied.) The model p.d.f. is sketched in figure 6.

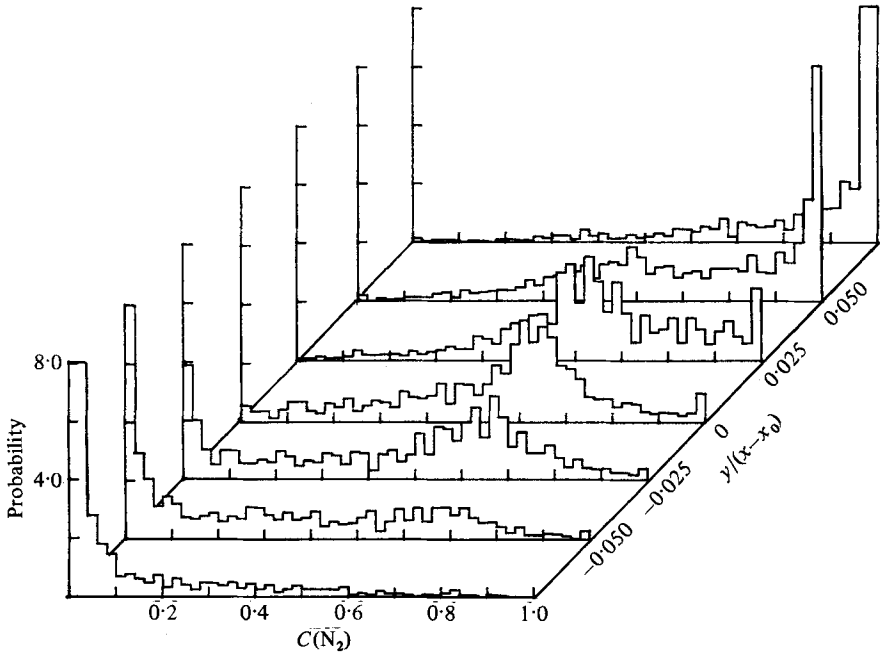


FIGURE 7. Probability density function; same condition as for figure 1 (*a*) (from Konrad 1976).

At this stage in our understanding of the fluid motions in the shear layer, the relative size of the four functions must come from experiment. The probe-resolution requirements are severe if the laminar diffusion zones are to be detected. Equations (7) and (8) indicate that for $Sc \sim 1$ the zone thickness is $\sim L/Re^{\frac{1}{2}}$ for the large-scale motions, where $\epsilon \sim \Delta U/L$ and $\sim L/Re^{\frac{3}{4}}$ for those near the Kolmogorov scale. These dimensions are such that even the high-resolution Brown probe used by Konrad would not resolve the strained layers under the conditions of his experiment. As a consequence, the probe indicates more molecular-scale mixing than actually occurs. (Breidenthal (1978) and Dimotakis (private communication) have noted this possibility.)

The p.d.f. deduced by Konrad from the concentration-probe measurements is given in figure 7. Before discussing a comparison of this figure with the model p.d.f., we note first that Konrad used the procedure of Toor (1962) to predict the amount of product that would be produced in a fast chemical reaction between reactants carried in the two streams at various equivalence ratios ϕ defined by

$$\phi \equiv \frac{\kappa_A(\infty)/\kappa_B(\infty)}{f}, \tag{9}$$

where f is the stoichiometric ratio of the chemical reaction between gases A and B carried in the two streams, and the κs are mass fractions. When we apply these results to the Wallace–Brown O_3–NO experiment, we find that the average temperatures exceed the experimental value in figure 4 by a factor of about two, a discrepancy that implies, indeed, an overestimate of molecular mixing from the probe data.

The conclusions we drew above from the Konrad experiment, namely, that there are large-scale zones of unmixed fluids throughout the layer and that lateral gradients in the molecularly mixed gases are small, should not be invalidated by this quantitative discrepancy.

The probe-resolution limitations do, however, cloud a comparison of figure 7 with the model p.d.f. The data contain high-probability peaks at the two free-stream concentrations and at the entrainment ratio. But whether these peaks are actually sharper than measured (only the values for pure A and B are expected to be delta functions) and whether thin laminar zones are present cannot be determined.

3. Scaling laws

To deduce the model scaling laws and the dependence of the amount of product on the various parameters, we now consider A and B to be reactive, and discuss the homogeneous zones and the strained laminar regions separately.

The rapid diffusion given by (6) shows that at sufficiently high Reynolds number the amount of product in the homogeneous zone is independent both of Re and Sc. It rises linearly with stoichiometric ratio ϕ until $\phi = E$, the entrainment ratio, above which condition it remains constant.

The amount of product in the flame sheet is the value per unit area multiplied by the total area, the latter quantity being independent of both Re and Sc.

The average product fraction per unit sheet area can be found from (7) and (8) to be

$$\bar{\kappa}_{\mathrm{p}} = \kappa_B(\infty)\left(\frac{2}{\pi}\frac{D}{\epsilon}\right)^{\frac{1}{2}}(\phi+1)\,e^{-\Lambda^2}, \tag{10}$$

where

$$\Lambda = \mathrm{erf}^{-1}\frac{\phi-1}{\phi+1}.$$

This expression can be written, using the strain rates at scales L and λ_0, as

$$\bar{\kappa}_{\mathrm{p}} = \kappa_B(\infty)\left(\frac{2}{\pi}\frac{1}{Sc\,Re}\right)^{\frac{1}{2}}L(\phi+1)\,e^{-\Lambda^2}, \tag{11}$$

$$\bar{\kappa}_{\mathrm{p}} = \kappa_B(\infty)\left(\frac{2}{\pi}\frac{1}{Sc}\right)^{\frac{1}{2}}\frac{L}{Re^{\frac{3}{4}}}(\phi+1)\,e^{-\Lambda^2} \tag{12}$$

respectively. Since most of the flame sheet is generated at the small scales, the latter expression is the more representative.

As already remarked, more experiments are needed before the relative contribution from the homogeneous region and the flame sheets can be determined. Some conclusions can, however, already be drawn. In water, with its small diffusion coefficient, $Sc \sim 1000$, the product in the flame sheet will be negligible for any reasonable ratio of homogeneous to flame-sheet volume. Therefore the total amount of product will be independent of Re and, for ϕ larger than E, independent of this parameter also.

In gases, for ϕ above E, the product amount will also depend upon Re and ϕ only in the flame sheets, with the total magnitude depending upon the relative amount of flame sheet.

Breidenthal (1978, 1982) made measurements of the product generation in a shear layer by a fast reaction in water using a technique that circumvents probe-resolution difficulties. He found that, for $\phi \gg 1$, the mean product thickness

$$P = \int \frac{\kappa_{\mathrm{p}}}{\kappa_B(\infty)}\,dy \tag{13}$$

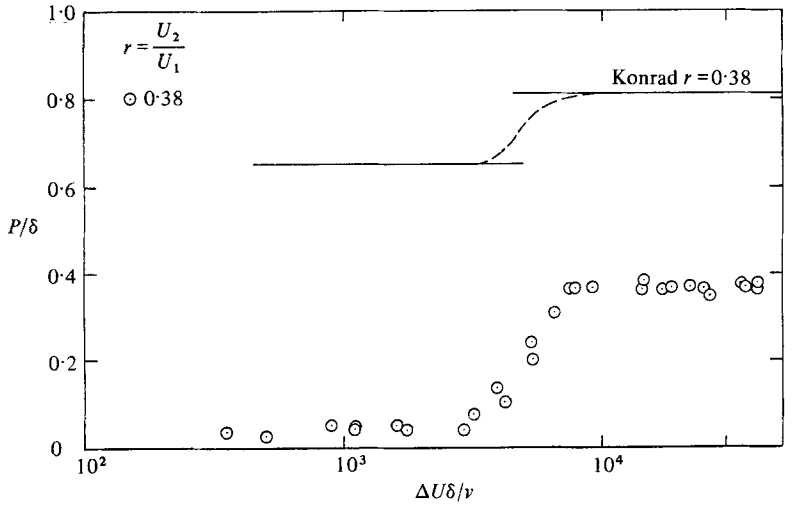


FIGURE 8. Dependence of mixing on large-scale Reynolds number for gases and liquids (from Breidenthal 1978, 1982).

was independent of Reynolds number above a critical transition value. The result is shown in figure 8, where P is normalized by the vorticity thickness

$$\delta = \frac{\Delta U}{(\partial U/\partial y)_{\max}}. \tag{14}$$

The Konrad data for gases also shown in this figure indicate the product amount to be independent of Re, but the probe limitations and the small Re-range make this result inconclusive. Note also that a reduction by a factor of two in the Konrad high-Reynolds-number value would make the gas and liquid results approximately equal, a result that implies that most of the product in gases is generated and resides in the homogeneous regions, i.e. that the relative amount of product in the flame sheets is small.

The scaling laws may be summarized as follows.

In liquids, all of the product resides in the homogeneous regions. The amount is independent of both Reynolds and Schmidt numbers, increases linearly with ϕ until $\phi = E$, and then remains constant.

The same description of the homogeneous region applies to gases. The amount of product in the flame sheets scales with the amount per unit area, since the flame sheet per unit volume is independent of Re, Sc, and ϕ. The product per unit flame sheet area is given in (11) and (12). The relative contribution from the two regions must be determined from additional experiments.

4. Discussion of related studies

No attempt will be made here at a discussion of all the literature containing ideas similar to those put forward above. Instead brief mention will be made only of those that seem especially pertinent.

The shortcomings of the elementary ideas of turbulent eddy diffusivity have long been recognized, especially for the treatment of chemical reactions. Hawthorne, Weddell & Hottel (1949), for instance, introduced the concept of 'unmixedness' and emphasized the need for an understanding of the molecular mixing processes. They

also formulate a model based on the idea that the combustion rate in flames is limited by the air entrainment rate.

The earliest work to separate entrainment and molecular mixing along the lines of the present paper was the study of hypersonic wakes by Proudian & Feldman (1965). Coherent structures had, of course, not been discovered at the time of that work, and the authors retain the concept of an interface between turbulent and non-turbulent fluid. What they propose, however, in common with the present model, is that fluid elements are transported, unmixed, into the wake by the large-scale turbulent motions, and mix after a lag time, which they similarly estimate from the cascade time. In this way they arrive at a two-component model for the wake structure, which they state leads to encouraging agreement with experimental observations.

The studies of turbulent diffusion flames, and especially the simulations of such flames by a chemical reaction in water (Hottel 1953), established that flame lengths in both circumstances were independent of Reynolds numbers at high Reynolds number. An attempt to explain this result led one of us (J.E.B. in Witte *et al.* 1974) to the idea of the two timescales and to the argument that, since the larger, controlling time was Reynolds-number independent, so was the flame length. The ideas were applied by Breidenthal (1978) in the interpretation of his shear-layer data, but the possible influence of the Reynolds and Schmidt numbers in gases, through their influence on the flame sheets, was overlooked.

It will be recognized that the present analysis has elements in common with the coherent flame-sheet model described in Marble & Broadwell (1977, 1980) – principally that a portion of the reaction takes place in strained laminar flames. It differs in two basic ways. The first, of course, is that these studies make use of the idea of gradient transport. The second is in the treatment of flame-sheet production and shortening. In these earlier works, the flame-sheet production rate was taken to depend on the mean strain rate, and shortening or flame sheet annihilation occurred when either fuel or oxidizer were totally consumed in local regions. In the present formulation, the argument that at high Reynolds number the annihilation by diffusion at the Kolmogorov scale is fast relative to flame-sheet production eliminates the need for the detailed balance equation of these earlier analyses.

The ESCIMO model of Spalding (1978) contains some of the physical concepts in the present formulation, but it is difficult at this stage in its development to make a more definite statement.

5. Conclusion

In the formulation described in §§2 and 3, the reaction between A and B was postulated to be fast relative to the mixing rate. This restriction was imposed to simplify the presentation and to obtain a few definite results. It should be clear that, if numerical computation were considered, then a chemical kinetic system with both fast and slow reactions could be treated. Here the restriction 'slow' means sufficiently slow that no reaction takes place in the flame sheets. If all the reactions are slow, the homogeneous region constitutes a 'well-stirred reactor', to which the free-stream constituents are steadily added.

The predictions of the proposed model and the differences from those of turbulent-diffusion models may be summarized as follows. First, according to the present view, the reaction between substances carried in the two streams occurs uniformly across the shear layer whatever the equivalence ratio ϕ. In diffusion analyses the reaction

zone is driven from one side of the layer to the other by changes in ϕ. Secondly the product flux has a markedly different dependence on ϕ in the two approaches. In our model with the concentration in one stream and the stoichiometric ratio fixed, an increase in ϕ above a critical value determined by the entrainment ratio causes more product to appear in the flame sheets but not in the homogeneously mixed regions. Therefore in liquids, where negligible reaction occurs in the sheets, no further product appears for ϕ larger than this critical value. In diffusion models, the product flux is monotonic with ϕ. Finally the molecular-diffusion coefficient plays a role in the proposed formulation but none in the usual approach.

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REFERENCES

Breidenthal, R. E. 1978 A chemically reacting, turbulent shear layer. Ph.D. thesis, Caltech.

Breidenthal, R. E. 1982 Structure in turbulent mixing layers and waves using a chemical reaction. *J. Fluid Mech.* **109**, 1.

Brown, G. L. & Rebollo, M. R. 1972 A small, fast response probe to measure composition of a binary gas mixture. *A.I.A.A. J.* **10**, 649.

Brown, G. L. & Roshko, A. 1974 On density effects and large structure in turbulent mixing layers. *J. Fluid Mech.* **64**, 775.

Chandrasada, C., Mehta, R. D., Weir, A. D. & Bradshaw, P. 1978 Effects of free stream turbulence on large structures in turbulent mixing layers. *J. Fluid Mech.* **85**, 693.

Coles, D. 1981 Prospects for useful research on coherent structure in turbulent shear flow. *Proc. Indian Acad. Sci.* (*Engng Sci.*) **4**, 111.

Corcos, G. M. & Sherman, F. S. 1976 Vorticity concentration and the dynamics of unstable free shear layers. *J. Fluid Mech.* **73**, 241.

Corrsin, S. 1974 Limitations of gradient transport models in random walks and in turbulence. *Adv. Geophys.* **18** A, 25.

Dash, S. M., Pergament, H. S. & Thorpe, R. O. 1979 A modular approach for the coupling of viscous and inviscid processes in exhaust plume flows. *A.I.A.A. Paper* no. 79-0150, *17th Aerospace Science Meeting, January* 1979.

Dimotakis, P. E. & Brown, G. L. 1976 The mixing layer at high Reynolds number: large structure dynamics and entrainment. *J. Fluid Mech.* **78**, 535.

Fiedler, H. F. 1974 Transport of heat across a plane turbulent mixing layer. *Adv. Geophys.* **18** A, 93.

Hawthorne, W. R., Weddell, D. S. & Hottel, H. C. 1949 Mixing and combustion in turbulent gas jets. In *Proc. 3rd Symp. on Combustion and Flame and Explosion Phenomena*, p. 266. Williams & Wilkins.

Hottel, H. C. 1953 Burning in laminar and turbulent fuel jets. In *Proc. 4th Symp.* (*Int.*) *on Combustion*, p. 97. Williams & Wilkins.

Konrad, J. H. 1976 An experimental investigation of mixing in two-dimensional turbulent shear flows with application to diffusion-limited chemical reactions. Ph.D. thesis, Caltech; and *Project SQUID Tech. Rep.* CIT-8-PU.

LANDAU, L. D. & LIFSHITZ, E. M. 1959 *Fluid Mechanics*. Pergamon.

MARBLE, F. E. & BROADWELL, J. E. 1977 The coherent flame model for turbulent chemical reactions. *Project SQUID Tech. Rep.* TRW-9-PU.

MARBLE, F. E. & BROADWELL, J. E. 1980 A theoretical analysis of nitric oxide production in a methane/air turbulent diffusion flame. *Interagency Energy/Environment R&D Program Rep.* EPA-600/7-80-018.

PATNAIK, P. C., SHERMAN, F. S. & CORCOS, G. M. 1976 A numerical simulation of Kelvin–Helmholtz waves of finite amplitude. *J. Fluid Mech.* **73**, 215.

PROUDIAN, A. P. & FELDMAN, S. 1965 A new model for mixing and fluctuations in a turbulent wake. *A.I.A.A. J.* **3**, 602.

REBOLLO, M. R. 1973 Analytical and experimental investigation of a turbulent mixing layer of different gases in a pressure gradient. Ph.D. thesis, Caltech.

SPALDING, D. B. 1978 A general theory of turbulent combustion. *Energy* **2**, 16.

TOOR, H. L. 1962 Mass transfer in dilute turbulent and nonturbulent systems with rapid irreversible reactions and equal diffusivities. *A.I.Ch.E. J.* **8**, 70.

UBEROI, M. S. & SINGH, P. I. 1975 Turbulent mixing in a two-dimensional jet. *Phys. Fluids* **18**, 764.

WALKER, B. J. 1979 Turbulence model comparisons for shear layers and axisymmetric jets. *U.S. Army Missile Command Tech. Rep.* RD-80-1.

WALLACE, A. K. & BROWN, G. L. 1979 A reacting shear layer with significant heat release: an experiment. In *Proc. 2nd Symp. on Turbulent Shear Flows, Imperial College, London, June* 1979.

WILLIAMS, F. A. & LIBBY, P. A. 1980 Some implications of recent theoretical studies in turbulent combustion. *A.I.A.A. J.* **19**, 261.

WITTE, A. B., BROADWELL, J. E., SHACKLEFORD, W. L., CUMMINGS, J. C., TROST, J. E., WHITEMAN, A. S., MARBLE, F. E., CRAWFORD, D. R. & JACOBS, T. A. 1974 Aerodynamic reactive flow studies of the H_2F_2 laser-II. *Air Force Weapons Lab., Kirtland Air Force Base, New Mexico* AFWL-TE-74-78, 33.